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OXYGEN SOURCES FOR FUEL CELLS

A. F. Forziati

D. Singman

25 April 1963



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FOR THE COMMANDER:  
Approved by



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## ABSTRACT

Commercially available gas, liquid, and chemical sources of oxygen for use with fuel cell batteries are compared. Cryogenic sources are shown to be the most efficient on a weight and volume basis. Chemical generators are satisfactory for applications requiring rather large quantities of oxygen gas at infrequent periods. Compressed gas cylinders are convenient when small quantities of oxygen are desired. A bibliography of selected publications during the past five years is included.

### 1. INTRODUCTION

The practicable utilization of present day fuel cells requires a dependable source of oxygen. The oxygen should be sufficiently pure that unimpaired operation of the battery is insured. The use of air directly is not considered in this survey as trace impurities would eventually impair the electrochemical efficiency of the oxygen electrode. Furthermore, the cell would be complicated by the addition of air pumps and chemical purification trains. For portable applications, the source and cell must be efficient on a weight and volume basis.

Available sources of oxygen consist of containers of gaseous or liquid oxygen and of chemical and mechanical generators. A comparison of these sources is needed in order to choose an oxygen supply offering the best compromise of lightness, convenience, and dependability.

### 2. SOURCES OF OXYGEN

#### 2.1 Compressed Gas

Compressed gas is commercially available in a variety of cylinder sizes, some of which are listed in table I. The 9 5/8-in. cylinders are not available separately but in stacks of 30, mounted on trailers. The stack is connected to a manifold so that the contents of the cylinders are delivered simultaneously. When the gas pressure drops to 50 lb/in.<sup>2</sup>, the valves are automatically closed and a signal notifies the operator to change trailers.

The weights of the cylinders of a given size may vary as much as 15 percent (1). This may amount to 10 lb for the 70 ft<sup>3</sup> and 20 lb for the 224 ft<sup>3</sup> sizes. Although the values for the ratio of the weight of gas to the gross weight are consequently approximate, it is still evident that use of the larger cylinders is more efficient from a weight standpoint.

The tabulated gross weights do not include weights of regulators required to reduce the gas pressure to the low values used by fuel cells. Such regulators weigh about 7 lb, which is less than the possible weight variance of the larger cylinders and, in these cases, may be neglected.

TABLE I. COMPARISON OF COMPRESSED OXYGEN CONTAINERS

Cylinder Dimensions		Cylinder volume <sup>b</sup>	Gross weight <sup>a</sup>	Gas volume <sup>a</sup>	Gas weight	Wt gas <sup>b</sup> gross wt	Vol gas <sup>b</sup> vol cylinder
diameter <sup>a</sup>	length <sup>a</sup>			(70°F, 1 atm)			
(in.)	(in.)	(ft <sup>3</sup> )	(lb)	(ft <sup>3</sup> )	(lb)	(%)	(ratio)
2	15	0.03	4	2	0.2	5	70:1
4	14	0.1	16	10	1	6	100:1
6	21	0.3	32	28	2	6	80:1
8	27	0.8	73	70	6	6	90:1
9	52	1.9	153	224	18	12	120:1
9 5/8	252	11.1	-	1540	128	-	140:1

<sup>a</sup>From ref 2<sup>b</sup>Calculated



## 2.2 Liquid Oxygen

Oxygen is also available in the liquid state. The containers vary in capacity from a few liters to several thousand gallons. The small containers are usually simple Dewar-type flasks made of glass or of spun copper. They depend on high vacuum and the reflectivity of the silvered glass or the polished copper surfaces for insulation. The containers are open to the atmosphere and the low temperature is maintained by the vaporization of liquid at a rate corresponding to the heat leakage. A 2-l flask may have an evaporation loss, called boil-off, of 6 percent of its contents per day (1).

Inexpensive moderate size containers are available for commercial uses requiring 6000 ft<sup>3</sup>, or more, of oxygen per month. These containers consist of an inner wall of polished stainless steel and a mild steel outer shell. The intervening space is filled with a mixture of inert powder and bright copper particles, then evacuated, and sealed. The cylinders hold about 26 gal of liquid oxygen under a pressure of 75 lb/in.<sup>2</sup> Excess pressure generated by oxygen boil-off is relieved through a vent valve. Thus with no additional equipment other than possibly a manifold, these cylinders can supply 3000 ft<sup>3</sup> of oxygen at 300 ft<sup>3</sup>/hr and 75 lb/in.<sup>2</sup> pressure. This is equivalent to 12 standard, 224 ft<sup>3</sup>, high-pressure cylinders (3).

Very efficient tanks for the storage and transportation of large quantities of liquid oxygen have been constructed. A 30,000-gal railway tank car is said to have an evaporation loss of only 0.1 percent per day (4). Such tanks employ high vacuum and multiple radiation shields to achieve the high degree of thermal isolation required. Table II lists data for Dewar-type flasks, pressurized liquid containers, and a selection of mobile and stationary tanks. It will be noted that once again, the larger tanks experience a smaller percentage boil-off and have a more favorable content to gross-weight or volume ratio.

Construction details of typical vacuum-jacketed storage vessels and descriptions of instrumentation for the control of liquid oxygen flow are available (10, 11, 12, 13, 14, 15). Storage containers and transfer equipment capable of safe operation by unskilled personnel and conforming to military specifications are in assembly-line production (16).

Progress in the state-of-the-art of cryogenic containers has been very rapid. It is believed that it will be possible to construct storage tanks with a gas to tank-volume ratio of 900:1 and a liquid to gross-weight ratio of 93:100 in the near future. Evaporation losses as little as 0.02 percent per day are anticipated. A number of contemplated containers are listed in table III.

The very large quantities of oxygen required at remote areas by the military have spurred the production of mobile and of assembled-on-the-site liquefying facilities. The capacities and weights of some of these units are given in table IV.

TABLE II. COMPARISON OF LIQUID OXYGEN CONTAINERS, PRESENT TECHNOLOGY

GROSS		CONTAINER		CONTENTS				RATIOS		LOSS	REMARKS	EXP.
Weight (lb)	Volume (l)	Weight (lb)	Gas volume at 70° F. (ft <sup>3</sup> )	Liquid volume at normal boiling point (ft <sup>3</sup> )	(gal)	Weight (lb)	Weight oxygen Gross weight (%)	Volume gas Volume container	Percent per day			
31	5	18	150	0.18	1.3	13	41	220:1	6			1
51	10	26	300	0.35	2.6	25	49	250:1	6			1
69	15	31	460	0.53	4.0	38	55	280:1	6		Hofman standard containers	1
110	25	49	760	0.88	6.6	63	56	320:1	5			1
200	50	77	1,500	1.8	13	130	62	340:1	3			1
420	100	165	3,000	3.5	26	250	60	400:1 <sup>a</sup>	3			1
760	200	255	6,100	7.1	53	500	66	430:1 <sup>a</sup>	3			1
463	12.4	215	3,000	3.45	26.1	248	53.6	240:1	1.85		LPC <sup>b</sup>	5
1,800	34	980	10,300	12	90	860	47	300:1	1.5		Vertical tank	6
2,270	51	760	18,400	21	150	1,510	66.6	360:1	0.8		Tank	6
6,710	198	1,960	57,400	67	500	4,750	70.8	290:1	1.0		Tank	6
12,700	245	5,200	90,600	105	780	7,500	59.1	370:1	1.0		Stationary tank	6
21,500	509	9,000	151,000	173	1,320	12,500	58.2	296:1	0.8		Tank on skids	6
52,00	840	12,000	480,000	560	4,200	40,000	77	380:1	1.1		LOX trailer	7,8
76,000	1,440	26,000	600,000	890	5,200	50,000	66	420:1	0.2		Horizontal tank	6
185,000	4,530	60,000	1,510,000	1,752	13,200	125,000	67.6	333:1	0.3		Stationary tank	6
341,000	3,740	90,700	3,000,000	3,500	26,000	250,000	73	800:1	0.45		Field erected tank	9
1,000,000	13,300	180,000	10,000,000	12,000	87,000	830,000	82	750:1	0.25		Field erected tank	9
2,490,000	34,400	390,500	25,000,000	29,000	220,000	2,100,000	84	720:1	0.15		Field erected tank	9

<sup>a</sup> Volume ratio calculated from dimensions of obrotund container.<sup>b</sup> Low pressure cylinder.

TABLE III LIQUID OXYGEN CONTAINERS, FUTURE TECHNOLOGY<sup>a</sup>

GROSS	CONTAINER	CONTENTS			RATIO		LOSS <sup>b</sup>	CONSTRUCTION <sup>c</sup>
Weight (lb)	Volume (ft <sup>3</sup> ) (lb)	Weight Volume	Gas volume at 1 atm, 700°F (ft <sup>3</sup> )	Liquid volume at normal boiling point (gal) <sup>b</sup>	Wt (lb)	Weight oxygen Gross weight (%)	Percent per day	
1,150	21 200		12,000 13	100	950	82 540:1	0.33	Aluminum, 1963
1,230	21 280		12,000 13	100	950	77 540:1	0.33	Steel, 1963
72,400	1000 5,400		810,000 940	7,000	67,000	93 780:1	0.08	Aluminum, 1963
75,700	1000 8,700		810,000 940	7,000	67,000	88 780:1	0.08	Steel, 1963
1,130	21 180		12,000 13	100	950	84 540:1	0.15	Aluminum & Steel, 1964
74,400	1100 7,400		810,000 940	7,000	67,000	90 720:1	0.03	Titanium & Steel, 1964
1,080	21 130		12,000 13	100	950	88 540:1	0.07	Reinforced Plastic, 1967
72,000	1100 5,000		810,000 940	7,000	67,000	93 720:1	0.02	Reinforced Plastic, 1967

<sup>a</sup> Calculated except where otherwise indicated.

**b Reference 7.**

**C<sub>1</sub> Projected technology for the year indicated.**

TABLE IV. CRYOGENIC OXYGEN GENERATING EQUIPMENT PRODUCED FOR THE MILITARY<sup>a</sup>

Name of Unit	Oxygen Capacity	Approximate Weight
USN shipboard plant	110 lb/hr	21,000 lb
Trailer-mounted generator	25 lb/hr	8,000 lb
Trailer-mounted generator	125 lb/hr	30,000 lb
Skid-mounted generator	83 lb/hr	23,000 lb
Field generator-semitrailer	5 tons/day	78,000 lb
Field operation-railway car	20 tons/day	-
Transportable generator	28.2 tons/day	-
Liquid oxygen facility	75 tons/day	-

<sup>a</sup>Reference 8.

TABLE V. COMPARISON OF SOLID CHEMICAL SOURCES

Source	Available Oxygen (wt-percent)	Gross wt of typical unit (lb)	Yield of typical unit (wt-percent)
Barium peroxide	9 <sup>a</sup>	-	-
Lithium peroxide	35 <sup>a</sup>	-	-
Sodium chlorate	34 <sup>b</sup>	88	15 <sup>c</sup>
Sodium peroxide	21 <sup>a</sup>	-	-
Potassium superoxide	32 <sup>c</sup>	4 <sup>d</sup>	15 <sup>d</sup>

<sup>a</sup>Stoichiometric yield

<sup>b</sup>Liberated by chlorate candle<sup>(22)</sup>

<sup>c</sup>Yield of commercial superoxide<sup>(21)</sup>

<sup>d</sup>Neither the weight of water needed for reaction nor the weight of control equipment is included.

<sup>e</sup>The yield is greater for additional candle furnaces per generator (23)

### 2.3 Chemical

Liquid oxygen is the most economical source of oxygen gas; it can not, however, be stored for long periods of time. The compressed gas, on the other hand, can be stored indefinitely but only in very limited amounts, within a reasonable volume. Chemical generators serve as a satisfactory compromise in applications requiring rather large quantities of oxygen gas at infrequent periods. Barium and sodium peroxides have been used to generate oxygen (17, 18, 19), lithium peroxide has been suggested (20), but the most successful chemical generators employ sodium chlorate or potassium superoxide. The amounts of oxygen available from these chemicals are compared in table V.

Potassium superoxide (21) is frequently used in face masks that do not admit air from the surrounding environment. The superoxide serves a dual purpose. Water vapor from the wearer's breath reacts with potassium superoxide forming potassium hydroxide and oxygen; the potassium hydroxide then reacts with the exhaled  $\text{CO}_2$  to form potassium carbonate. Thus the superoxide not only generates oxygen but also removes  $\text{CO}_2$  from the system. As the amount of oxygen generated is somewhat greater than is required for normal breathing, the excess is vented to the outside through a one-way valve.

Self-regulating generators furnishing moderate amounts of oxygen on demand have been built commercially (21). A simple oxygen generator (fig. 1) consisting of three compartments, one above the other, may be readily assembled. Potassium superoxide is placed in the middle chamber and water in the top. Water flows down and fills the lower compartment eventually contacting the superoxide in the middle. The oxygen gas generated forces the water back up and stops further reaction. When gas is withdrawn, the drop in pressure (in the middle chamber) causes water to again enter the middle chamber and oxygen generation is resumed. This arrangement could be utilized to furnish oxygen for a hydrogen/oxygen fuel cell; the water required for the decomposition of the  $\text{KO}_2$  would be obtained from the cell.

Sodium chlorate yields slightly more oxygen, by weight, than does potassium superoxide (34% for  $\text{NaClO}_3$  vs 32% for  $\text{KO}_2$ ). The equipment required for oxygen generation is somewhat more complicated (22). Sodium chlorate is mixed with iron powder, barium peroxide, and an inorganic binder and fabricated into candles. When ignited by means of a percussion cap, phosphorus match, or electric squib, the candle burns and liberates oxygen at a rate proportional to the burning area for a period determined by the length of the candle. Oxidation of the iron powder to iron oxide furnishes heat to continue decomposition of the chlorate; the barium peroxide combines with any chlorine that may be liberated during the reaction.

Although chlorate candles appear to have unlimited shelf life, once ignited, the candle is generally burned to completion; excess oxygen is stored under pressure in reserve tanks. Hence sodium chlorate is known as

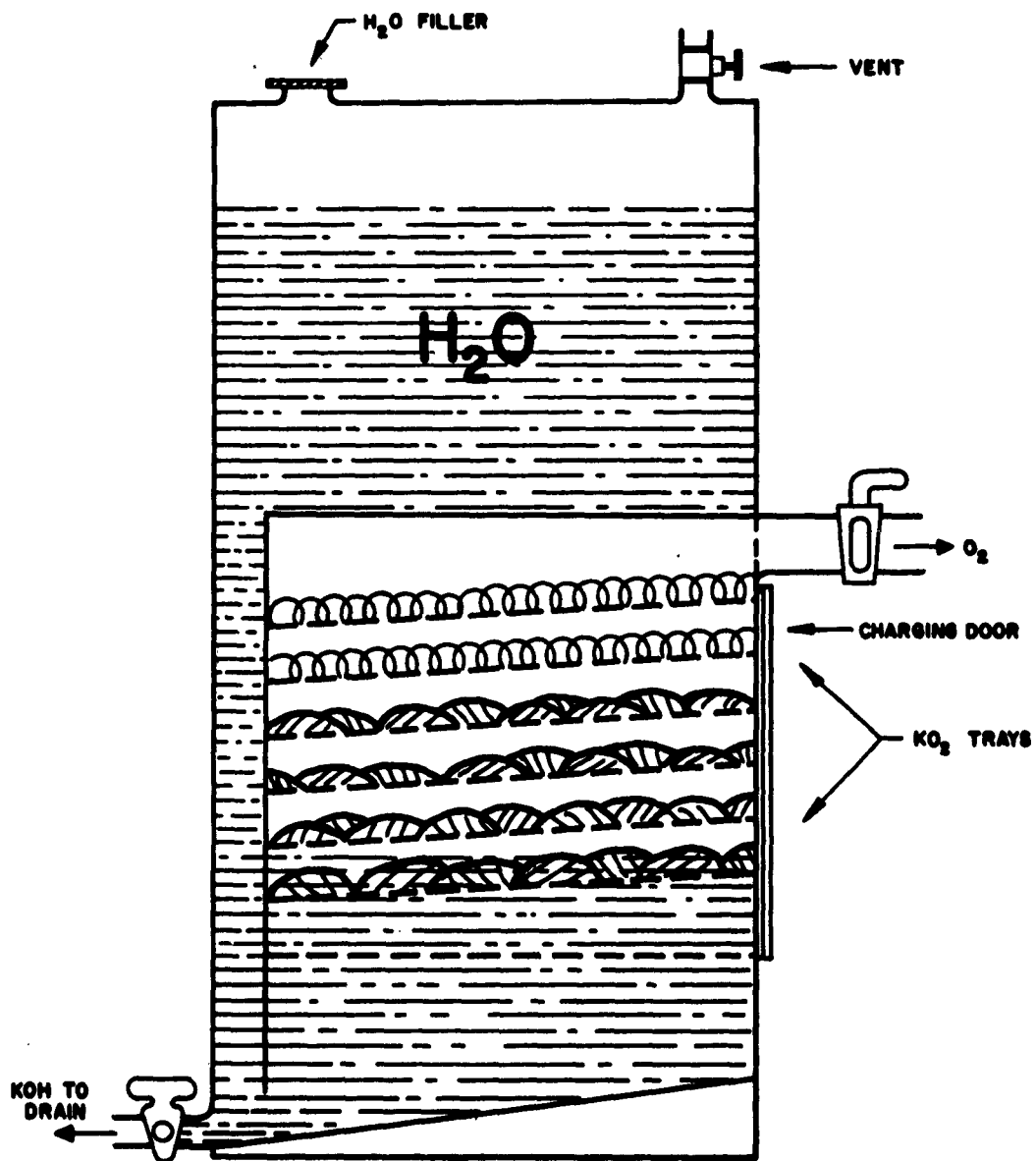


Figure 1. "Demand" oxygen generator.

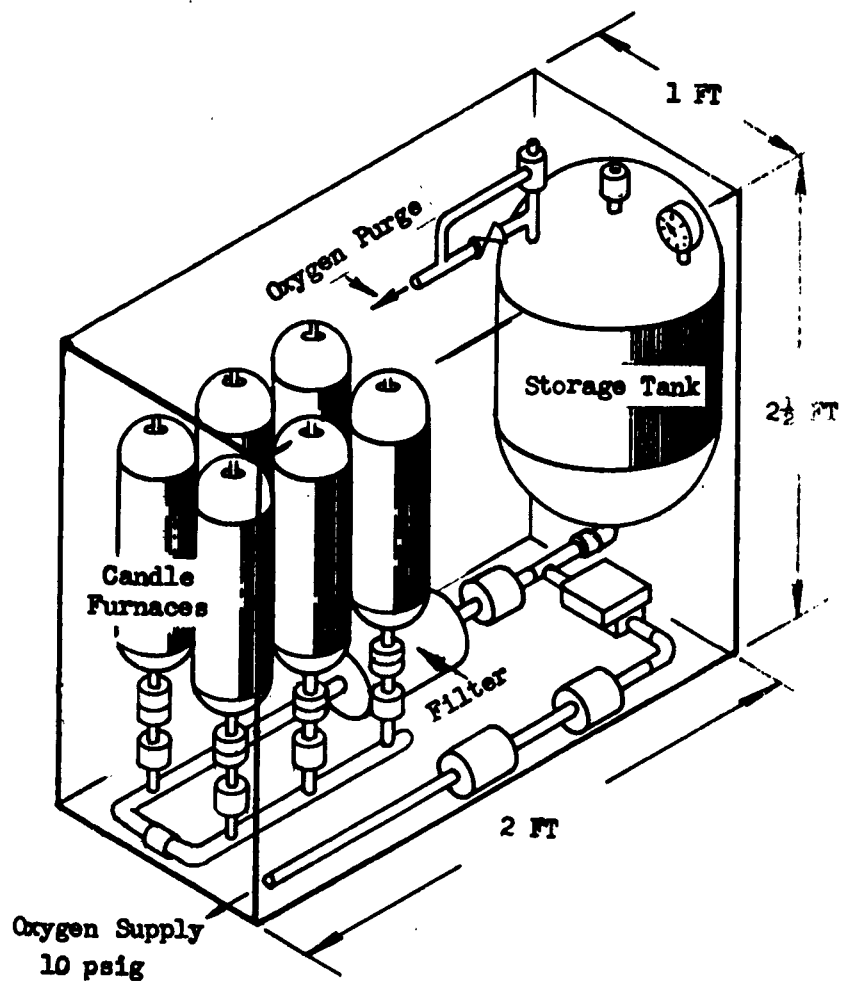
a controlled oxygen source; that is, the size and shape of the candle determine the rate and period of oxygen generation. Potassium superoxide, on the other hand, can generate oxygen on demand and within limits, in proportion to the demand. It is therefore considered to be a demand chemical source of oxygen.

A sodium chlorate oxygen generator is available commercially (21). The unit consists of a chlorate candle burner, two storage tanks capable of withstanding an internal pressure of 400 lb/in.<sup>2</sup>, and a reducing valve outlet. About 35 ft<sup>3</sup> of oxygen are generated by one candle. A check valve permits changing candles without loss of oxygen stored in the tanks. By attaching a number of burners to a manifold and firing the candles electrically, one at a time, whenever the pressure drops to a preset value, a continuous oxygen supply may be attained. A unit comprising six candles, a storage tank, check valves, pressure switches, and firing mechanisms has been designed (fig. 2) (23). Its weight and volume are estimated at 88 lb and 5 ft<sup>3</sup>, respectively. This unit can furnish oxygen for about 6 1/2 hr at 25 ft<sup>3</sup>/hr and 10 psig, or a total of 162 ft<sup>3</sup> over a longer period. The weight efficiency of the unit (ratio of weight of gas to weight of unit) is about 15 percent for one set of six candles. As the equipment can be used indefinitely, the weight efficiency of the system may be increased by providing additional sets of candles, for example, one additional set raises the weight efficiency to 21 per cent. The weight efficiencies of some chemicals and units used to generate oxygen are compared in table V.

The oxygen from any one of the three sources discussed above is satisfactorily pure for fuel cell use. Compressed "extra dry grade" oxygen is stated to have a minimum purity of 99.6 percent (2). Cryogenic oxygen is 99.5 to 99.6 percent pure; the major part of the impurity is argon (24, 25). Oxygen from a chlorate generator is about 99.5 percent pure (22). It contains small amounts of sodium chloride smoke and traces of carbon dioxide and carbon monoxide. Less than 0.05 percent of carbon dioxide and 0.007 percent of carbon monoxide, by volume, are present. The superoxide generator produces oxygen described as breathing grade. Oxygen obtained by the electrolysis of water has a purity of 99.5 percent (24).

### 3. ELECTROCHEMICAL POWER FROM OXYGEN SOURCES

A hydrogen/oxygen fuel cell, operating at 100 percent efficiency, yields about 1.9 kw-hr/lb and about 0.16 kw-hr/ft<sup>3</sup> of oxygen consumed. To assist in the selection of oxygen supplies for such cells, the amounts of power that may be generated by the oxygen available from the various units are listed in table VI. Power per pound and per cubic foot of source are also shown. Although present oxygen electrode efficiencies are about 75 percent, 100-percent efficiency was assumed in calculating the data tabulated; the weights and volumes of regulating equipment were neglected.



**Figure 2. Controlled oxygen generator.**



TABLE VI. ELECTROCHEMICAL POWER FROM VARIOUS OXYGEN SOURCES<sup>a</sup>  
(Hydrogen/Oxygen Cell)

Source	kw-hr per unit	kw-hr per lb of unit	kw-hr per ft <sup>3</sup> of unit
A Compressed gas cylinders			
1. Smallest size	0.3	0.08	10
2. Largest size	35	0.24	18
B. Liquid containers			
1. Smallest Dewar	24	0.78	35
2. Largest Dewar	940	1.2	67
3. Low-pressure cylinder	460	1.0	38
4. Tank - 51 ft <sup>3</sup> volume	2,800	1.2	55
5. Tank - 198 ft <sup>3</sup> volume	8,900	1.3	45
6. Tank - 245 ft <sup>3</sup> volume	14,000	1.2	57
7. Tank - 509 ft <sup>3</sup> volume	23,000	1.1	46
8. Tank - 4,530 ft <sup>3</sup> volume	230,000	1.3	52
9. LOX Trailer	75,000	1.6	89
C. Solid generator			
Chlorate	25	0.28	5
Superoxide	1.1	0.22	16

<sup>a</sup>Values are computed for an oxygen electrode assumed to be 100% efficient.  
Weights and volumes of regulatory equipment are not included.

The power generated by oxygen in other electrochemical systems will be different from the values tabulated. The ratios of the amounts of power generated by the oxygen from the various sources will be independent of the particular system employed.

#### 4. SUMMARY

A graphical comparison of the various oxygen supplies is shown in figure 3. Compressed-gas cylinders and solid-chemical generators possess the best shelf life as there is no loss of oxygen due to evaporation. The weight efficiency of compressed-gas cylinders, however, is only 5 to 15 percent because the containers must be made of heavy steel to withstand high pressures, about 2000 psi. The weight efficiency of chlorate generators is limited by the 34 weight percent yield of the chemical mixture. Systems for handling liquid oxygen in large, insulated, double-walled vessels have been developed. These containers are much lighter because their thin walls need withstand pressures of only a few pounds above atmospheric, thus weight efficiencies as high as 84 percent may be achieved.

The volume ratio of gas at atmospheric pressure to gas at 2,000 psi is approximately 130:1. One volume of chlorate candle yields about 300 volumes of gaseous oxygen. However, one volume of liquid oxygen produces 860 volumes of gas. Therefore, transportation and storage of oxygen in the liquid form is most economical on a volume as well as a weight basis.

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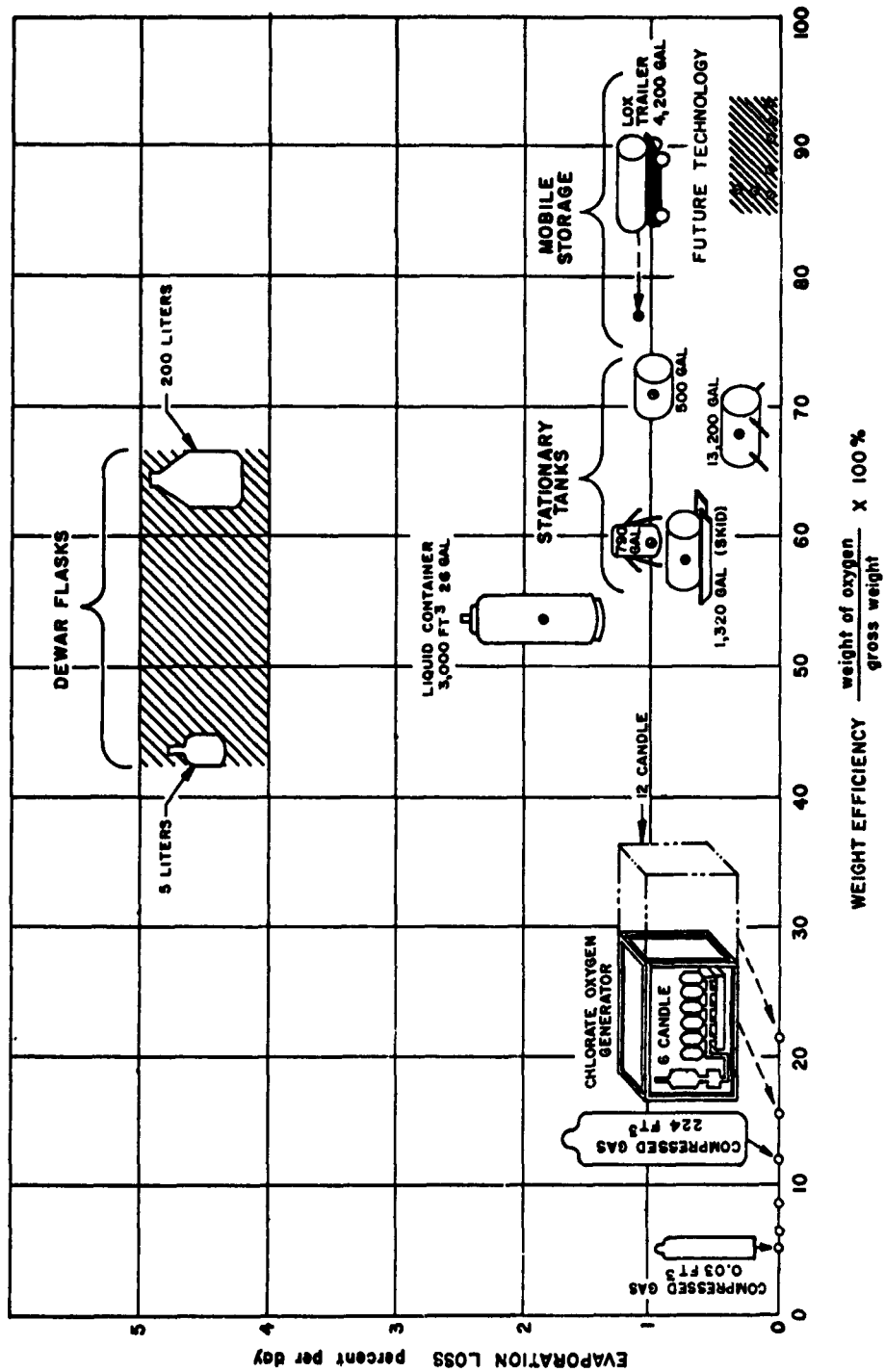


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